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PULSE SHAPING BASED ULTRA-BROADBANDWIDTH MULTIDIMENSIONAL SPECTROCOPIC METHODS

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14. ABSTRACT
Single-beam approaches to coherent anti-Stokes Raman scattering (CARS) have been developed to allow diagnostic capabilities to study molecular systems under extreme conditions such as those found in combustors. Unlike methods that combine beams with different wavelengths at different angles, the approaches developed use a single beam, thus simplifying implementation outside the research laboratory. Collaboration with scientists at the Air Force Research Laboratory (Wright-Patterson AFB) resulted in six publication and one manuscript pending submission. The first confocal images of flames providing species selective density and temperature were recorded.

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Laser spectroscopy, temperature and density measurements in flames.

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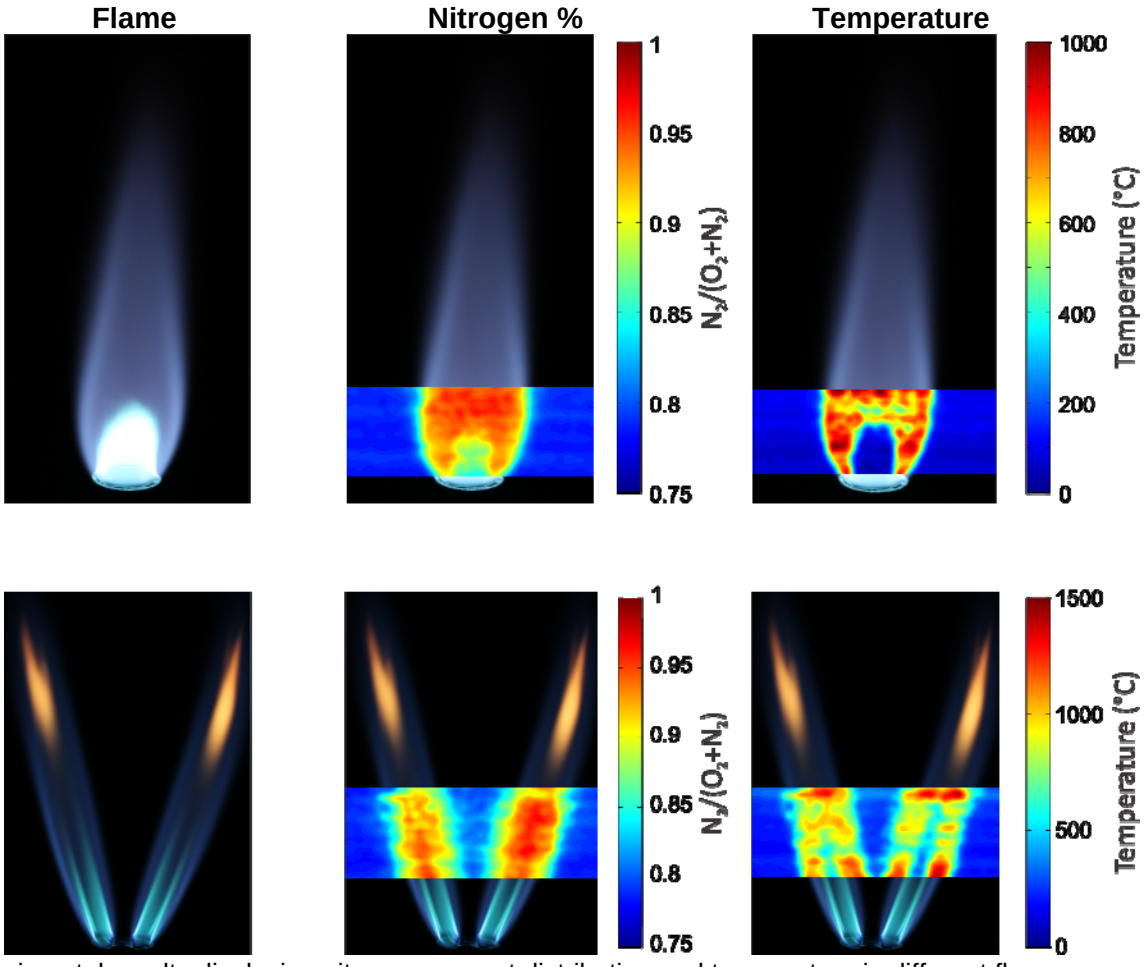
Objective: Develop a greatly simplified approach to coherent anti-Stokes Raman scattering (CARS), allowing us to extend its diagnostic capability to study molecular systems under extreme conditions. Unlike methods that combine beams with different wavelengths at different angles, our approach uses a single beam, thus simplifying measurements on difficult to reach locations such as within reactive flows. Collaborate with scientists at the Air Force Research Laboratory (Wright-Patterson AFB) where temperature and pressure measurements in reactive flows such as those of aircraft turbines are required.

Accomplishments:

1. Flame diagnostics working closely with AFRL scientists:

Accomplished confocal imaging of flames to provide species specific density and temperature information using single-beam CARS.



Experimental results displaying nitrogen percent distribution and temperature in different flames.

2. **Refining single-beam temperature determination and calibration**:
Figure 1. Experimental measurement of CARS vibration-rotational spectra for oxygen at room temperature (left) and high temperature (right). (red and blue) The rotational features are simulated based on a Boltzmann distribution of populations for each temperature. Notice that in addition to changes in the rotational distribution the vibrational hot-band is observed at high temperatures. The vibrational hot band provides additional temperature information based on it expected Boltzmann population. A temperature and pressure controlled cell from the AFRL has arrived at MSU, and we are using it to calibrate our method.

3. **New probe methods (narrower, multiple, and broader) for improved signal and multiplexing.**
Instead of using one broad-bandwidth pump and one narrow bandwidth probe, we decided to explore the use of multiple probe frequencies as shown in the next figure.

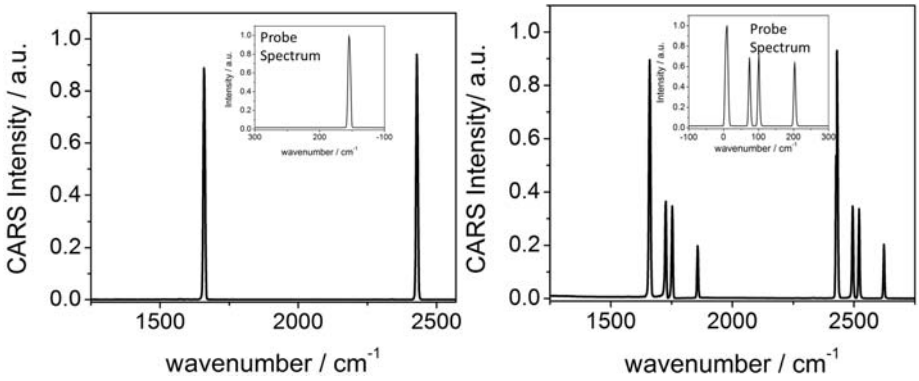


Fig. 2. (left) Single beam CARS spectrum of air (oxygen and nitrogen) obtained with a single frequency probe (see insert). (right) Single beam CARS spectrum of air, this time obtained with a probe beam that has four discrete frequencies. The four frequencies lead to four lines for each of the two species. While this complicates the spectrum, the pattern is easy to discern and provides higher signal to noise data.

3. **Single-shot species selective single beam CARS**.
The multiple probe wavelengths, when generated in a pulse shaper, can each receive a calibrated delay, and therefore can be used to provide the coherence dephasing time, a measure that is sensitive to temperature, directly in a single laser shot. This approach led to accurate temperature determination in a single laser shot and to the following publication.

O. Yue, M. Bremer, D. Pestov, J. R. Gord, S. Roy, and M. Dantus, "Gas Phase Thermometry via Multi-Time-to-Frequency Mapping of Coherence Dephasing", J. Phys. Chem. A, (2012)
In this paper we demonstrated a single-shot method for measuring the species-selective concentration and temperature. The advantages of this method are illustrated in the figures below.

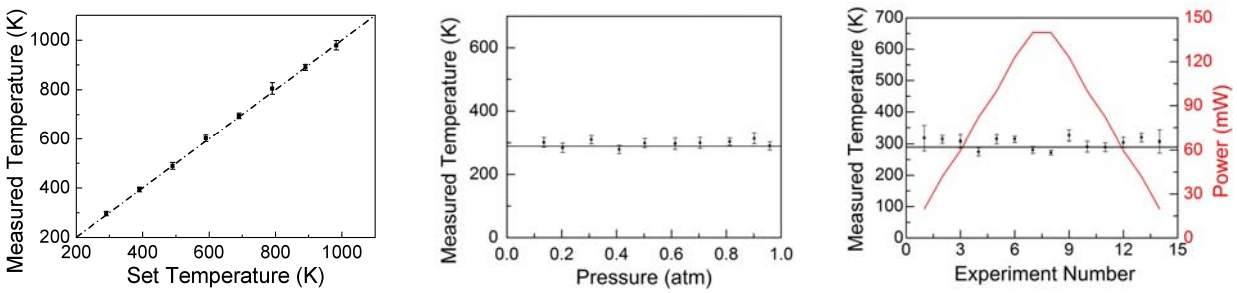


Fig. 3 (left) The measurements are calibrated and show precision and accuracy of 25K. (Middle) The method is independent of changes in pressure/density. This is very important because nanosecond laser based methods are very sensitive to pressure/density changes because collisions occur within the time of the measurement. In this method the measurement is completed within a couple of picoseconds and therefore there is no influence from pressure/density. (right) Because the measurement is "ratiometric" it is independent of laser intensity fluctuations. In this figure the average power of the laser is changes from 20-140mW (a factor of 7) and yet the temperature variations registered are within 10% of the target temperature. We are presently using this approach to image a propane/air flame.

Results from this Grant:

Peer Reviewed Journal Articles:

"Species-Selective Confocal flame imaging," A. van Rhijin, O. Yue, M.T. Bremer, J.R. Gord, S. Roy, and M. Dantus, Nature Photonics (to be submitted September 2013).

"Single-Shot Gas-Phase Thermometry by Time-To-Frequency Mapping of Coherence Dephasing," O. Yue, M.T. Bremer, D. Pestov, J.R. Gord, S. Roy, and M. Dantus, J. Phys. Chem. A 116, 8138–8141, (2012).

"Binary Phase Shaping for Selective Single-Beam CARS Spectroscopy and Imaging of Gas-Phase Molecules," P.J. Wrzesinski, D. Pestov, V.V. Lozovoy, B. Xu, S. Roy, J.R. Gord, and M. Dantus, J. Raman Spectrosc. 42 (3), 393–398 (2011).

"Group-Velocity-Dispersion Measurements of Atmospheric and Combustion-Related Gases using an Ultrabroadband-Laser Source," P.J. Wrzesinski, D. Pestov, V.V. Lozovoy, J.R. Gord, M. Dantus, and S. Roy, Opt. Express 19 (6), 5163–5170 (2011).

"Single-Beam CARS Imaging for Reacting Flow Diagnostics," P.J. Wrzesinski, D. Pestov, V.V. Lozovoy, S. Roy, J.R. Gord, and M. Dantus, Optics and Photonics News 21 (12), 49 (2010).

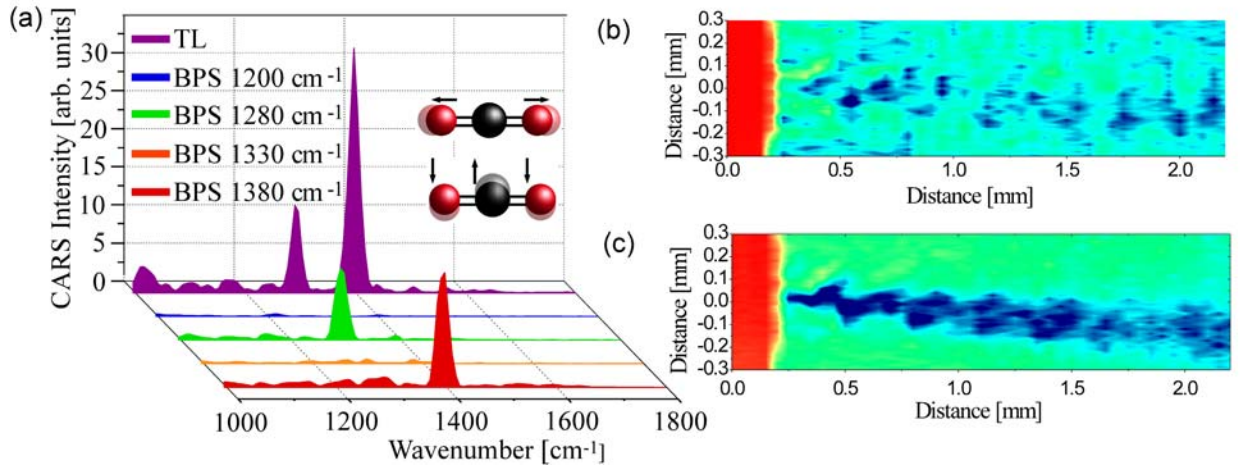


Figure 2 (from the OPN article). Being able to image reacting flows requires selective excitation of desired Raman vibrational transitions. (left) Selective excitation of the Fermi diad in CO_2 associated with the symmetric stretch and two quanta of the bending motion. Note that selective excitation at different frequencies is capable of isolating single vibrational transitions. (right) Single beam CARS imaging of a turbulent flow of CO_2 molecules. The top image is obtained with transform limited pulses, the bottom image is obtained with selective excitation of the symmetric stretch.

"Single-Beam Coherent Anti-Stokes Raman Scattering (CARS) Spectroscopy of Gas-Phase CO2 via Phase and Polarization Shaping of a Broadband Continuum," S. Roy, P.J. Wrzesinski, D. Pestov, M. Dantus, and J.R. Gord, J. Raman Spectrosc. 41 (10) 1194–1199 (2010).

"Single-Beam Coherent Anti-Stokes Raman Scattering Spectroscopy of N2 using a Shaped 7 fs Laser Pulse," S. Roy, P.J. Wrzesinski, D. Pestov, T. Gunaratne, M. Dantus, and J.R. Gord, Appl. Phys. Lett. 95 (7), 074102 (2009).

Invited Talks:

"Phase Control of Laser-Matter Interactions and Applications" **M.** Dantus, Committee on Atomic, Molecular, and Optical Sciences, April 3-4, 2012, National Academies' Keck Center –Washington, DC

"Pulse Shaping Based Ultra-Broadbandwidth Multidimensional Spectroscopic Methods," M. Dantus AFOSR Contractors Meeting, January 5-6, 2012

"Improved Axial Resolution for Single-Beam CARS Imaging," P.J. Wrzesinski, D. Pestov, V.V. Lozovoy, S. Roy, J.R. Gord, and M. Dantus, 10th European Conference on Nonlinear Optical Spectroscopy, May 22-25, 2011, Enschede, Netherlands.

"Imaging reactive flows and trace quantities of hazardous substances using single beam CARS"P.J. Wrzesinski, D. Pestov, V. V. Lozovoy, S. Roy, J.R. Gord, and M. Dantus, PQE Snowbird, Utah (Jan, 2011)

"Single-Beam Femtosecond CARS Spectroscopy of N2 and CO2 with an Ultrafast Supercontinuum," S. Roy, P.J. Wrzesinski, D. Pestov, M. Dantus, and J.R. Gord, 41st Central Regional Meeting of the American Chemical Society Poster Session, June 16-19, 2010, Dayton, OH.

"Pulse Shaping for Single-Beam CARS Spectroscopy and Imaging," P.J. Wrzesinski, D. Pestov, V.V. Lozovoy, B. Xu, S. Roy, J.R. Gord, and M. Dantus, 41st Central Regional Meeting of the American Chemical Society, June 16-19, 2010, Dayton, OH.

"Single-Beam CARS Spectroscopy with Ultrashort Laser Pulses for Applications Involving Limited Optical Access," J.R. Gord, P.J. Wrzesinski, D. Pestov, M. Dantus, and S. Roy, 2010 Augmentor Design Systems Conference, March 17-19, 2010, Jacksonville, FL.

"Single-Beam CARS Spectroscopy of Gas-Phase N2 and CO2," P.J. Wrzesinski, D. Pestov, M. Dantus, S. Roy, and J.R. Gord, Nonlinear Optics at 50 Symposium, October 26, 2011, Ann Arbor, MI.

"Propulsion Measurements with Single-Beam fs-CARS," J.R. Gord, D. Pestov, M. Dantus, P.J. Wrzesinski, and S. Roy, 7th Annual Dayton Engineering Sciences Symposium, October 24, 2011, Dayton, OH.

Contributed Presentations:

"Thermometry of Flames using Multiple Probe Single Beam CARS Spectroscopy," O. Yue, M.T. Bremer, D. Pestov, J.R. Gord, and M. Dantus, Conference on Lasers and Electro-Optics/Quantum Electronics and Laser Science Conference (CLEO/QELS), May 6-11, 2012, San Jose, CA.

"Single-Beam CARS Spectroscopy of Gas-Phase N_2 and CO_2," P.J. Wrzesinski, D. Pestov, M. Dantus, S. Roy, and J.R. Gord, Nonlinear Optics at 50 Symposium, October 26, 2011, Ann Arbor, MI.

"Propulsion Measurements with Single-Beam fs-CARS," J.R. Gord, D. Pestov, M. Dantus, P.J. Wrzesinski, and S. Roy, 7th Annual Dayton Engineering Sciences Symposium, October 24, 2011, Dayton, OH.

"Single-Beam CARS Imaging for Reacting Flow Diagnostics," P.J. Wrzesinski, D. Pestov, V.V. Lozovoy, S. Roy, J.R. Gord, and M. Dantus, Conference on Lasers and Electro-Optics/Quantum Electronics and Laser Science Conference (CLEO/QELS), May 1-6, 2011, Baltimore, MD.

"Single Beam fs-CARS for Selective Excitation and Measurements of Group-Velocity Dispersion," J.R. Gord, P.J. Wrzesinski, D. Pestov, M. Dantus, and S. Roy, 36th Annual Dayton-Cincinnati Aerospace Science Symposium, March 1, 2011, Dayton, OH.

"Towards Single-beam CARS Imaging of Reacting Flows" P.J. Wrzesinski, D. Pestov, V. V. Lozovoy, S. Roy, J.R. Gord, and M. Dantus, Frontiers in Optics Rochester, NY (October, 2010): Talk

"Pulse Shaping Strategies for Single-Beam CARS," P.J. Wrzesinski, V.V. Lozovoy, D. Pestov, S. Roy, M. Dantus, and J.R. Gord, 22nd International Conference on Raman Spectroscopy, August 8-13, 2010, Boston, MA.

"Strategies for Selective and Multiplexed Single Beam CARS Imaging," P.J. Wrzesinski, D. Pestov, V.V. Lozovoy, S. Roy, J.R. Gord, and M. Dantus, 9th European Conference on Nonlinear Optical Spectroscopy, June 21-23, 2010, Bremen, Germany.

"Single-Beam Femtosecond CARS Spectroscopy of N_2 and CO_2 with an Ultrafast Supercontinuum," S. Roy, P.J. Wrzesinski, D. Pestov, M. Dantus, and J.R. Gord, 41st Central Regional Meeting of the American Chemical Society, June 16-19, 2010, Dayton, OH.

"Prospects and Challenges of Single-Beam CARS Spectroscopy with Sub-10-fs Laser Pulses," J.R. Gord, P.J. Wrzesinski, D. Pestov, M. Dantus, S. Roy, 35th Annual Dayton-Cincinnati Aerospace Science Symposium, March 9, 2010, Dayton, OH.

"Single-Beam CARS Spectroscopy with Sub-10-fs Laser Pulses," J.R. Gord, P.J. Wrzesinski, D. Pestov, M. Dantus, and S. Roy, Joint Poster Session of the Dayton Section American Chemical Society and the Ohio Valley Section Society for Applied Spectroscopy, March 8, 2010, Dayton, OH.

"Single-Beam Coherent Anti-Stokes Raman Scattering (CARS) Spectroscopy with Tailored Ultrashort Laser Pulses," J.R. Gord, P.J. Wrzesinski, D. Pestov, T. Gunaratne, M. Dantus, and S. Roy, AIAA-2010-1398, 48th AIAA Aerospace Sciences Meeting, January 4-7, 2010, Orlando, FL.

"Single-Beam CARS Spectroscopy of N_2 and CO_2 using an Ultrashort Laser Pulse," J.R. Gord, P.J. Wrzesinski, D. Pestov, T. Gunaratne, M. Dantus, and S. Roy, 5th Annual Dayton Engineering Science Symposium, October 26, 2009, Dayton, OH.

"Development of a Novel Femtosecond Laser Machining/Drilling Technology Based on Temporal Pulse Shaping and In-Situ Inspection Methods," S. Gogineni, S. Roy, M. Dantus, and J.R. Gord, 5th Annual Dayton Engineering Science Symposium, October 26, 2009, Dayton, OH.

"Single-Beam CARS Spectroscopy of Gas-Phase N_2 and CO_2," S. Roy, P.J. Wrzesinski, D. Pestov, M. Dantus, and J.R. Gord, Gordon Research Conference on Laser Diagnostics in Combustion, August 16-21, 2009, Waterville Valley, NH.

"Femtosecond Coherent Anti-Stokes Raman Scattering (fs-CARS) Spectroscopy of N_2 with a Shaped 7-fs Laser Pulse," S. Roy, P.J. Wrzesinski, D. Pestov, T. Gunaratne, M. Dantus, and J.R. Gord, Joint Poster Session of the Dayton Section American Chemical Society and the Ohio Valley Section Society for Applied Spectroscopy, March 3, 2009, Dayton, OH.

"Single-Beam Coherent Anti-Stokes Raman Scattering (CARS) Spectroscopy of N_2 using a 5-fs Laser Pulse," S. Roy, P.J. Wrzesinski, D. Pestov, T. Gunaratne, M. Dantus, and J.R. Gord, 34th Annual Dayton-Cincinnati Aerospace Science Symposium, March 3, 2009, Dayton, OH.

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